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DRY FALLOUT OF SEA-SALT PARTICLES AND ITS SEASONAL AND DIURNAL VARIATION

By

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Abstract

A dry fallout gauge is devised, which can determine dry fallout of the sea-salt particles, by the use of Farlow's halide-ion sensitive film, in the daytime as well as at night. The ten-day-average rate of dry fallout of sea-salt particles is determined in Kyoto for three years since November, 1962. The rate of fallout is high in winter, low in summer, and nearly the same feature is observed for three years. This seems to be caused by a larger supply of the particles from the Sea of Japan in the winter season. The total dry fallout of sea salt in Kyoto is found to be 1.7×10^{-5} gm cm^{-2} year^{-1}, and the average size distribution of the particles is determined. It is also found that there is a conspicuous diurnal variation in the rate of dry fallout: the value is several times larger at night than in the daytime. This is caused by diurnal variations in relative humidity and number concentration of the particles in the lowest atmosphere. The fact that the rate of dry fallout is only a small per cent of the total ground sink of sea-salt particles is noticed, and a discussion is given.

1. Introduction

The sea-salt particles in the atmosphere are produced on the sea surface, as a result of the direct interation between the atmosphere and the ocean, and form an important portion of the atmospheric condensation nuclei. Especially, giant sea-salt particles act in producing the first precipitation elements in non-freezing clouds. The present study is a part of a series of the authors' study on production of the particles on the sea surface, distribution in the atmosphere, and elucidation of the role of the particles in the precipitation mechanism.

The particles produced on the sea surface are carried over land by turbulent diffusion and wind. On land, there is a large ground sink consisting of the dry fallout and the impaction by trees and other ground obstacles. It is necessary to determine the amount of the ground sink in order to establish the particle distribution in the atmosphere over land. This was one aim of the present study.

The sea-salt particles make up the principal constituent of atmospheric salt, and the determination of the ground sink of particles is very important, also from the geochemical point of view, as the problem of movement of substance from the ocean to the continent.

Since there seems to be a sharp reduction of the number concentration, in the lowest a few hundred meters, caused by the ground sink (Toba, 1965a), the ground observation of the sea-salt particles is subject to a considerable handicap ; nevertheless, the ground observation is expected also to afford useful information for the study of variation in the number concentration or of the role of the particles in precipitation processes, as the ground observation is much easier to make compared with the observation in the sky, and may accumulate continuous observational data.

Toba and Tanaka (1963) made an observation of sea-salt particles which fell to the ground in Kyoto, every night for one month in the season of winter monsoon, and at several stations along a transversal cross-section of Japan in the direction of the monsoon on a few nights, and it was shown to some extent that number concentration of the particles was related to wind speed over the Sea of Japan, and to the distance inland.

A dry-fallout gauge was devised for the present study, which could be operated in the daytime as well as at night, and the ten-day-average rate of dry fallout of giant sea-salt particles for various mass ranges of the nuclei was determined for three years. The reason why we chose the ten-day average was with the intention of finding some clue for the understanding of precipitation mechanism, rather from an analysis of the rate of fallout and the type of salt-mass distribution of the particles, which might correspond to the average meteorological conditions in the ten days, than from a study of the phase of the fluctuating number concentration. Also, a determination of the number concentration for each range of the mass of salt of the particles was made by an impactor, near ground at several stations including Kyoto, every afternoon for the first one year in the above period. The results of the observation by the impactor will be reported elsewhere. In this article, a study by use of the dry-fallout gauge is mainly reported.

2. Method of the observation—a dry-fallout gauge for sea-salt particles

(a) *Sampling surface*

The sampling surface for sea-salt particles used in the present study is the same as that used by the authors in 1963, i.e., a halide ion-sensitive film prepared by the method of N. H. Farlow (1954 and 1957). The method of preparation and treatment of the film was described by Toba and Tanaka (1963).

The white spots developed on the film were measured, in the present study, from 100-time enlarged prints of microscopic pictures by use of a scale specially prepared for the measurement. The relation between the mass of chloride contained in the particles and the size of the white spot on the film was again strictly calibrated by the use of an isopiestic method.

(b) *Dry-fallout gauge and a comparison with other methods of the observation*

In 1963, the authors used a small can, with the sampling film at the bottom, to measure the dry fallout of sea-salt particles. Since the sampling film is, however, slightly photosensitive, this method may be used only at night. A newly devised dry-fallout gauge for the present study is shown in Fig. 1. The roof protects the film from precipitation, and the under surface and the inside wall of the cylinder in the center are painted black, so that daylight is almost absorbed by the black surfaces before it reaches the film which is set at the bottom of the cylinder. Since the terminal velocity of the particles is much smaller compared to that of the precipitation, the particles are not intercepted by the roof so long as some movement of air exists. By this method the film may be continuously exposed to the air for ten days or so with no

Fig. 1. The standard-type dry-fallout gauge for sea-salt particles.

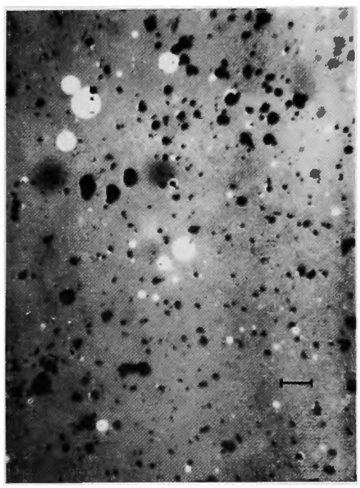

Fig. 2. Example of the sampling surface with white circular spots developed by sea-salt particles. The largest spot corresponds to 1.3×10^{-9} gm in the mass of salt, and black images are soot or some other materials. The scale indicates 100μ.

appreciable change in color. The dimension of the gauge is 50 cm in both height and diameter of the roof, 4 cm in diameter of the opening of the cylinder. The distance between the roof and the opening of the cylinder is variable, and 20 cm has been usually used. Fig. 2 shows an example of the sampling surface which showed white spots by the fallout of sea-salt particles.

In order to know the meaning of the observational values by the dry-fallout gauge, a comparative observation was made by three different methods at the same time. A sampling film was set in the gauge, and the other film was nakedly held horizontally near the gauge at the same height as the opening of the gauge, and both films were exposed for 12 hours, 6 : 00 p.m. on August 3 through 6 : 00 a.m. on August 4. The observation by the gauge was made for another 12 hours, 6 : 00 a.m. through 6 : 00 p.m. on August 4. At the same time, number concentration of the particles was measured by use of the impactor, and the relative humidity by an Assman's aspiration psychrometer, every one hour during the above 24 hours. Full lines in Fig. 3 illustrate the average rate of dry fallout of sea-salt particles (F) obtained by the gauge, a dash-dot line shows the values obtained by the naked film, and dashed lines indicate the average values of the products of the number con-

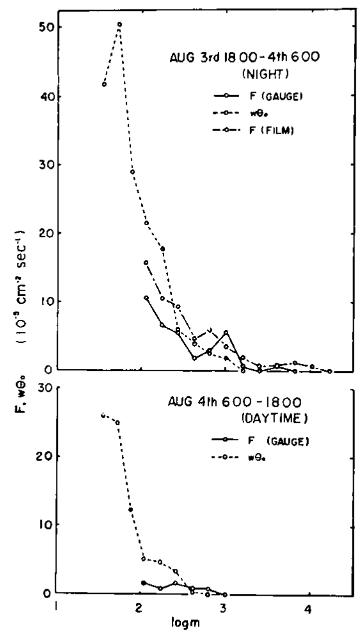

Fig. 3. A comparison among observations of the rate of dry fallout (F) by the dry-fallout gauge and by a naked sampling film, and an observation of the product of terminal velocity and number concentration ($w\theta_0$) by an impactor. (See the context.)

centration (θ_0), and the terminal velocity (w) of the particles, which was calculated from the value of relative humidity and the mass of salt of the particles. The upper part of Fig. 3 is for the 12 hours at night, and the lower part for the 12 hours in the daytime. In both parts of the figure, the curves have practically similar values, although the values of $w\theta_0$ for smaller values of $\log m$ (m: mass of salt in a particle in 10^{-12} gm unit) seem to be higher than those of the F and the values by the naked film are slightly higher than that by the gauge. One thing should be noted here. In Fig. 3, values for $w\theta_0$ are entered up to $\log m=1.45$, while values for F are entered only up to $\log m=1.97$. The reason is that the reading of the white spots on the film in the observation by the impactor was made by the microphotographic method; while, in the observation of F in this test experiment, the reading was made by use of a reading microscope, since the absolute number of the particles to be read on the film was small, and the lower limit of the measurement by the reading microscope was found to be about $\log m=1.95$. It may be concluded here that, for particles larger than about $m=10^{-10}$ gm, the value of the rate of dry fallout (F) obtained by the dry-fallout gauge approximately indicates the average of the product of w and θ_0.

In order to further examine whether there is some difference in the measured values of F due to the difference in the shape and dimension of the gauge, another dry-fallout gauge was made as shown in Fig. 4. The diameter of the cylinder is the same as the standard type, but the roof is much smaller and closer to the opening of the cylinder. Fig. 5 shows a comparison of

Fig. 4. The chimney-type dry-fallout gauge for sea-salt particles.

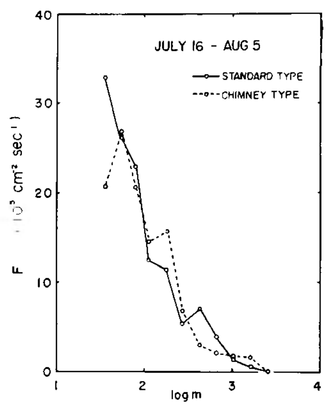

Fig. 5. A comparison of the observational values of F by the standard-type and the chimney-type dry-fallout gauges. (July 16 through August 5, 1965.)

the obtained average values of F with the two types of the gauge for 20 days. The curves effectively coincide with each other, and this indicates that the effect of the shape of the gauge is insignificant as long as average values are concerned. Under conditions, however, such as thunderstorms and typhoons, rain often immerses the film in the chimney-type gauge and the film is ruined. Consequently, the standard type gauge is better for the routine observation.

3. Dry fallout of sea-salt particles in Kyoto

(a) *Seasonal variation*

In Fig. 6 is illustrated the ten-day-average rate of dry fallout of sea-salt particles in Kyoto, obseved by the use of the standard-type dry-fallout gauge, for three years since November 20, 1962. There is a general trend of the seasonal variation which is common for three years, i.e., the rate of fallout is higher in winter and lower in summer: nearly three quarters of the total annual fallout fall during six months October through March. This trend seems to mainly correspond to the northwesterly winter monsoon which brings more sea-salt particles from the Sea of Japan.

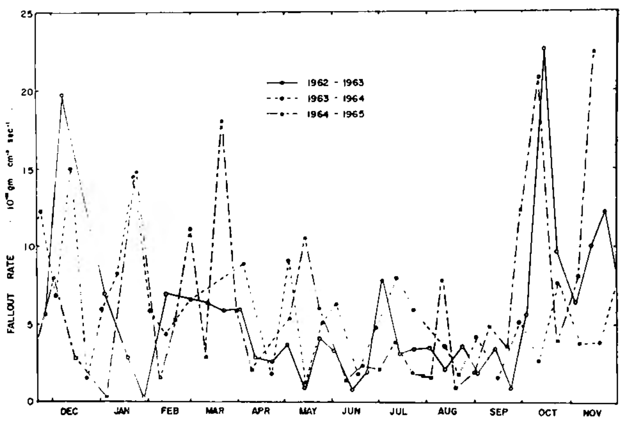

Fig. 6. Seasonal variation of the ten-day average rate of dry fallout (unit: 10^{-13} gm cm^{-2} sec^{-1}) of sea-salt particles larger than 2.8×10^{-11} gm in the mass of salt. (November, 1962 through November, 1965.)

(b) *Total fallout rate of sea salt and particle size distribution*

In Fig. 7 are illustrated the frequency distributions of the dry fallout of the particles and of the salt mass for each range of the mass of salt contained in the particles. In the calculation of the mass of salt, it is assumed that each particle has the composition of sea salt. The smaller the mass of salt, the larger the value of F, whereas there is a maximum in the

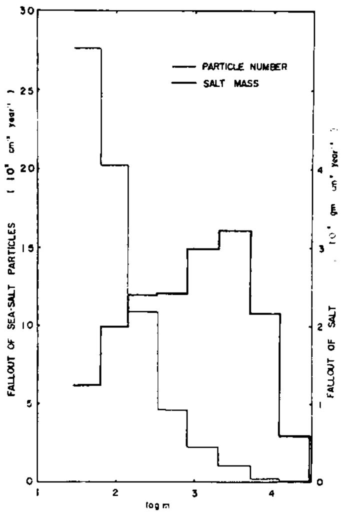

Fig. 7. Frequency distributions of the dry fallout of sea-salt particles (10^3 cm^{-2} year^{-1}) and of the dry fallout of salt (10^{-6} gm cm^{-2} year^{-1}) for each range of the mass of salt contained in the particle.

value of the mF between 10^{-9} gm and 10^{-8} gm in the mass of salt, and particles smaller than $\log m = 1.45$ do not significantly contribute to the rate of fallout of salt. The total fallout of sea salt was 1.60×10^{-5} gm cm^{-2} year^{-1} for the first year, 1.83×10^{-5} gm cm^{-2} year^{-1} for the second year, and 1.65×10^{-5} gm cm^{-2} year^{-1} for the third year.

(c) *Comparison with results of the observation by the impactor*

In most of the first year, observation of the surface concentration of sea-salt particle number (θ_0) was made every day in the afternoon. From this

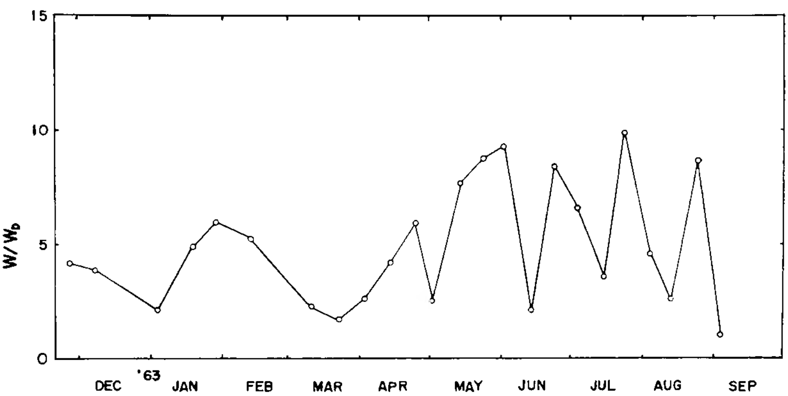

Fig. 8. Ratio between $F/\theta_0(=w)$ and the terminal velocity of dry sea-salt nuclei (w_D) for the range of salt mass between $\log m = 2.15$ and 2.53. (November, 1962 through September, 1963, see the context.)

and the above mentioned ten-day average rate of fallout (F), and by use of the relation obtained in 2: $F=w\theta_0$, the ten-day average of apparent velocity of gravitational fall of the particles (w) is calculated for some ranges of the mass of salt. In Fig. 8 is illustrated the ratio between the w and the terminal velocity of dry particles (w_D), for a range of the mass of salt of $\log m = 2.15$ through 2.53. The value is always higher than 1, the maximum being 10; and the average value is 4.9. The ratio, w/w_D is approximately a function only of relative humidity (RH) as was shown by Toba (1965b). Since the value of $w/w_D=4.9$ corresponds to RH of 97%, and the relative humidity in the afternoon is usually much lower, the above value of w/w_D presupposes a diurnal variation of θ_0 and/or RH. The diurnal variation really exists as will be shown in the next section.

4. Diurnal variation of the concentration and dry fallout of sea-salt particles

Fig. 9 is the time-series representation of results of the observation of

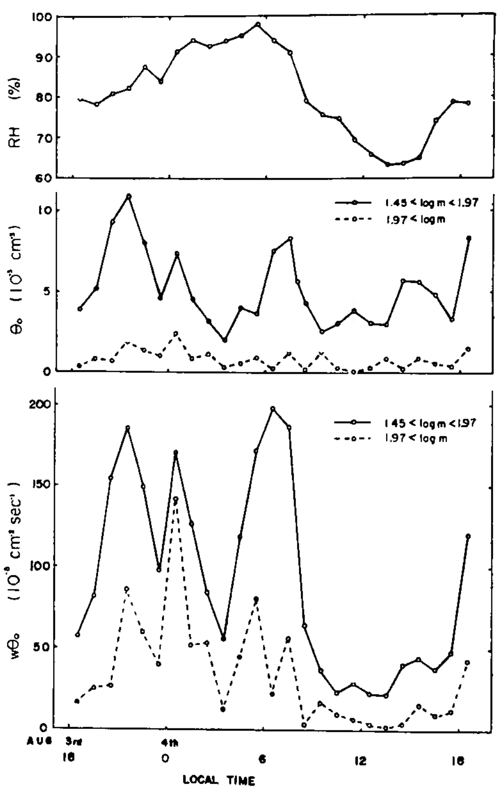

Fig. 9. Diurnal variation of relative humidity (RH), number concentration (θ_0) and the rate of fallout ($w\theta_0$), of sea-salt particles. (6:00 p.m. August 3 through 6:00 p.m. August 4, 1965.)

diurnal variation of relative humidity (RH), number concentration of sea-salt particles (θ_0), and the rate of dry fallout of the particles $(w\theta_0)$ which was once mentioned in 2(b). The observation was made every one hour beginning 6:30 p.m. August 3, 1965 for 24 hours. The weather was typical summer, and was constant for several days. The relative humidity was high at night and reached the maximum of 98% about sunrise, and decreased in the daytime, the minimum value being 63% in the afternoon. The values of θ_0 and $w\theta_0$ are shown for two ranges of the mass of salt: larger than and smaller than $\log m=1.97$ $(m=9.4\times10^{-11}$ gm). There are two maxima in these values at about 9:00 p.m. and at about 8:00 a.m. The diurnal variation of $w\theta_0$ is much larger than that of θ_0, and the rate of dry fallout at night is several times higher than that in the daytime. The fact that the value of the $w\theta_0$ for the particle range of larger masses of salt is especially high between 9:00 p.m. and 2:00 a.m. relatively to the value for the range of smaller masses of salt, seems to demonstrate that larger particles rapidly fall out as the relative humidity increases at night. Salt-mass distributions of the average rate of fallout of the particles during the 12 hours at night and the 12 hours in the daytime calculated from this observation were shown in Fig. 3, and they were compared with the result of the observation of the rate of dry fallout (F), as was described in 2. The average rate for 24 hours is several times higher than the afternoon values, and this is the explanation of the value of w/w_D which is shown in Fig. 8.

This is an observation only of one day. The finding, however, is found more universal from the result of the following five-day observation. The observation of θ_0 and RH were made at 3:00 p.m. everyday, August 1 through August 5, 1965. At the same time, the dry fallout was observed by two standard-type dry-fallout gauges: one was open for all days and the other for 6 hours 12:00 through 6:00 p.m., for the 5 days: 6:00 p.m. July 31 to 6:00 p.m. August 5, 1965, the above mentioned twenty-four-hours observation being made on the fourth of the five-day observation. The results are illustrated in Fig. 10 and Fig. 11. The humidity in the afternoon was between 65% and 76%, the weather was actually constant, and there was no extreme change in the particle concentration. The values of F and $w\theta_0$ have practically similar values for particles larger than $\log m=2$ for the afternoon observation, but the value of F obtained by the all-day gauge is much larger than that by the afternoon gauge.

In Fig. 12 is shown a comparison of the fallout rates by the afternoon gauge and by the all-day gauge during a period of July 16 through July 26, 1965. The value by the all-day gauge is utmost two times larger than that by the afternoon gauge. Since there was an intermittent rain throughout

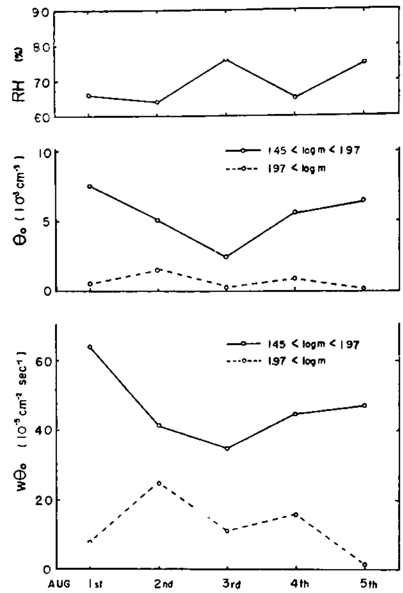

Fig. 10. Five-day observation of relative humidity (RH), number concentration (θ_0) and the rate of fallout ($w\theta_0$), of sea-salt particles. (August 1 through August 5, 1965.)

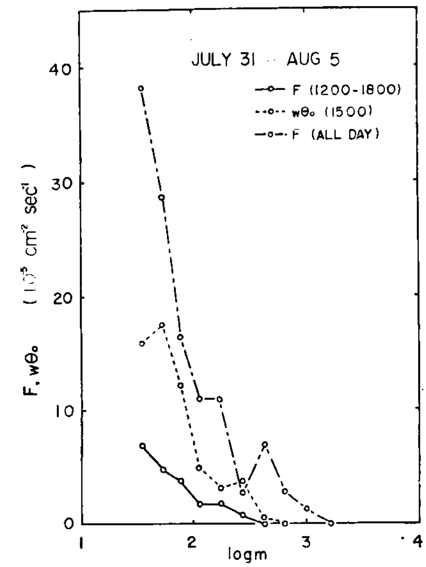

Fig. 11. A comparison among the rates of dry fallout (F) by the all-day gauge, by the afternoon gauge, and the afternoon values of $w\theta_0$ by the impactor measurement. (6:00 p.m. July 31 through 6:00 p.m. August 5, 1965.)

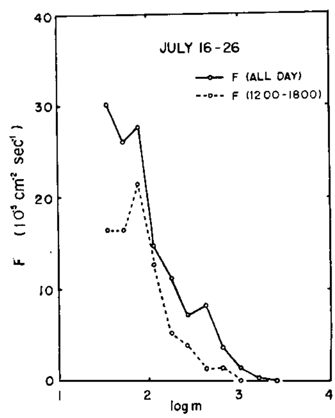

Fig. 12. A comparison between the rates of dry fallout (F) by the all-day gauge, and by the afternoon gauge. (July 16 through July 26, 1965.)

this period, the reason is inferred as there was no significant diurnal variation in relative humidity in this period.

5. Concluding remarks

As a result of the present study, the following items have been made clear.

(1) The dry-fallout gauge may well be used to determine the time integral of the value of $w\theta_0$: a product of the number concentration (θ_0) of sea-salt particles near the ground and the terminal velocity (w) of the particles, for the particles larger than 10^{-10} gm in the mass of salt.

(2) The difference of the observed values due to shape of the gauge does not seem to be very significant.

(3) The total dry fallout of sea salt in Kyoto is 1.7×10^{-5} gm cm^{-2} year^{-1}, and the average size distribution of the particles has also been determined (Fig. 7).

(4) The feature of the seasonal variation of the dry fallout in Kyoto has been found (Fig. 6).

(5) As to the diurnal variation, the amount of the fallout at night is several times larger than in the daytime, and it is caused by the diurnal variation of relative humidity and the surface concentration of the particles.

There are many fluctuations, in Fig. 6, of values of the rate of dry fallout besides the main trend of the seasonal variation. Also, some different types of the particle size distribution were observed. It will be interesting to study these fluctuations and the type of size distribution in relation to meteorological conditions that prevailed in each ten days. However, this is not included in the present report.

Before the present study had been done, we expected that the gauge might determine the total ground sink of sea-salt particles, but the result is that it determines only the term $w\theta_0$: the dry fallout of the particles. The ground sink consists of the term $w\theta_0$ and the term $(D\partial\theta/\partial z)_{z=0}$; the latter is the amount of particles brought down by turbulent diffusion due to vertical gradient of the particle concentration, which seems to be caused by the impaction of the particles by trees and other ground obstacles. According to Toba (1965a), the latter term is much more important in the total ground sink, and this concept is supported by the results of the present study. Namely, the value of dry fallout of 1.7×10^{-5} gm cm^{-2} year^{-1} corresponds to the concentration of salt in natural water of 0.17 mg liter^{-1}, when the salt dissolves into rain water of 10^3 mm year^{-1}. The usual salt concentration of river water is several mg liter^{-1}, and that of rain water is a few mg liter^{-1}. So, the total ground sink of the particles will correspond to the concentration of a few mg liter^{-1}. Consequently, the term $w\theta_0$ is to be of the order of one twentieth or one thirtieth of the total ground sink of the particles. It should be noted now that we should distingwish the dry fallout from the total ground sink, and that we cannot directly determine the total ground sink by the measurement of the dry fallout of the particles.

Novertheless, there is a possibility that we may relate the value of θ_0 with the term $(D\partial\theta/\partial z)_{z=0}$, as was treated by Toba (1965a). The establishment of this relation, however, necessitates a further study of the vertical gradient of the particle concentration $(\partial\theta/\partial z)$ as well as the eddy diffusivity (D), in the lowest several hundred meters. This line of study is left for the future.

Acknowledgments

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